

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**

## WEST

## End of Result Set

 [Generate Collection](#) [Print](#)

L12: Entry 1 of 1

File: DWPI

Sep 20, 1994

DERWENT-ACC-NO: 1994-338145

DERWENT-WEEK: 199442

COPYRIGHT 2002 DERWENT INFORMATION LTD

TITLE: Impregnating metal in carbon@ material, for slip rings etc. - by heat-treating carbon@ material and impregnating metal in material at temp. higher than m.pt of metal

PATENT-ASSIGNEE: NIPPON STEEL CHEM CO (YAWH), NIPPON STEEL CORP CO (YAWA)

PRIORITY-DATA: 1993JP-0077522 (March 12, 1993)

## PATENT-FAMILY:

PUB-NO	PUB-DATE	LANGUAGE	PAGES	MAIN-IPC
JP <u>06263571</u> A	September 20, 1994	.	004	C04B041/88

## APPLICATION-DATA:

PUB-NO	APPL-DATE	APPL-NO	DESCRIPTOR
JP06263571A	March 12, 1993	1993JP-0077522	

INT-CL (IPC): B60L 5/00; C04B 35/52; C04B 41/88

ABSTRACTED-PUB-NO: JP06263571A

## BASIC-ABSTRACT:

Impregnating metal comprises heat-treating the C material at 900-1200 deg.C, and impregnating the metal in the material at a temp. 30-400 deg.C higher than the m.pt. of the metal but less than 1200 deg.C.

USE - For making slides or slip rings.

ABSTRACTED-PUB-NO: JP06263571A

## EQUIVALENT-ABSTRACTS:

CHOSEN-DRAWING: Dwg. 0/0

DERWENT-CLASS: L02 L03 M22 Q14 V04 X12 X23

CPI-CODES: L02-H04; L02-J01; M22-G03K;

EPI-CODES: V04-L01A; X12-G02; X23-A03;

# PATENT ABSTRACTS OF JAPAN

(11)Publication number : **06-263571**  
(43)Date of publication of application : **20.09.1994**

---

(51)Int.CI. **C04B 41/88**  
**B60L 5/00**  
**C04B 35/52**

---

(21)Application number : **05-077522** (71)Applicant : **NIPPON STEEL CORP**  
**NIPPON STEEL CHEM CO LTD**  
(22)Date of filing : **12.03.1993** (72)Inventor : **FUJIMOTO KENICHIRO**  
**MUKAI KOICHIRO**

---

## (54) METHOD FOR IMPREGNATING CARBON MATERIAL WITH METAL

### (57)Abstract:

PURPOSE: To improve the impregnation of a metal into a carbon material by impregnating the carbon material fired at a specific temperature with the metal melted at a higher temperature than the melting point of the metal in a nonoxidative atmosphere.

CONSTITUTION: A porous carbon material produced from petroleum pitch coke is subjected to a degassing treatment at 900-1200°C for 30min to 1-30hrs in a nonoxidative atmosphere such as Ar. A metal selected from copper, copper alloy, aluminum, zinc, lead, etc., in response to the purpose is charged in a high pressure container, thermally melted at a temperature 30-400°C higher than the melting point of the metal but lower than 1200°C, and subsequently mixed with the treated carbon material under a high pressure to impregnate the carbon material with the melted metal. The carbon material is immersed in the melted metal for a prescribed time, discharged and subsequently cooled to provide the carbon-metal composite material having a high metal impregnation degree.

---

### LEGAL STATUS

[Date of request for examination]  
[Date of sending the examiner's decision of rejection]  
[Kind of final disposal of application other than the examiner's decision of rejection or application converted registration]  
[Date of final disposal for application]  
[Patent number]  
[Date of registration]  
[Number of appeal against examiner's decision of rejection]  
[Date of requesting appeal against examiner's decision of rejection]  
[Date of extinction of right]

Copyright (C); 1998,2003 Japan Patent Office

\* NOTICES \*

**Japan Patent Office is not responsible for any damages caused by the use of this translation.**

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

---

## DETAILED DESCRIPTION

---

### [Detailed Description of the Invention]

[0001]

[Industrial Application] this invention relates to the method of carrying out melting sinking in of the metal into the detailed pore of a carbon material, and manufacturing composite material. Furthermore, if it says in detail, it is related with the metal sinking-in method of making it performing good to the carbon material calcinated below at metal sinking-in processing temperature to metal sink in.

[0002]

[Description of the Prior Art] Recently, the composite material sunk in and obtained by the carbon material in a metal attracts attention as the charge of sliding current collection material, a sliding material, etc.

[0003] Usually, the method of lowering pulling up a carbon material from molten metal, and holding a pressurization state, after having carried out the temperature up, having carried out melting of the metal, raising the pressure in a high pressure vessel with the state where the carbon material was immersed into molten metal and performing fixed time pressurization sinking-in processing, while sinking-in processing of the metal at the time of manufacturing the above composite material performed deaeration processing of a carbon material, after preparing the metal and the carbon material in the high pressure vessel is taken.

[0004] Moreover, in order to make it easy metaled to sink in, being set as a remarkable elevated temperature is more common the temperature which carries out melting sinking in of the metal ] than the melting point of the metal which sinks in so that a metaled fluidity may be raised and viscosity can be lowered.

[0005] However, in spite of it not being necessarily easy to fully be filled up with a metal into the detailed pore of these carbon materials and being the pore connected on the surface of the carbon material, the present condition is that the portion into which a metal does not fully sink has occurred not a little.

[0006] For this reason, the thing for which the solution of a metal salt is beforehand sunk in and heat-treated to (1) carbon material as a method of aiming at sufficient sinking [ of the metal to the pore in a carbon material ] in, and a solvent is removed, Methods, such as using, after the wettability to (2) carbon materials which improves the wettability to the carbon material of molten metal (Provisional-Publication-No. No. 136644 [ 61 to ] official report) heat-treats (3) carbon materials using a good alloy (Japanese Patent Publication No. No. 822 [ 52 to ] official report) in the temperature region more than the temperature of baking carbonization (publication-number No. 83784 [ four to ] official report), are proposed.

[0007]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the method of sinking in the metal of sufficient amount for the pore in the carbon material calcinated below at metal sinking-in processing temperature.

[0008] Moreover, another purpose is sinking in a metal, without raising the degree of graphitization of the carbon material used as a base material not much.

[0009]

[Means for Solving the Problem] By suppressing heat treatment temperature, this invention persons found out that the aforementioned purpose could be attained, and completed this invention while they heat-treated and did degasifying of the carbon material in advance of metal sinking-in processing and suppressed generating of the cracked gas from the carbon material under metal sinking-in processing.

[0010] After this invention heat-treats more than for 30 minutes under a non-oxidizing atmosphere beforehand within the limits of 900 degrees-C or more less than 1200 degree-C temperature to the carbon material calcinated below at the metal sinking-in processing temperature used as a raw material, it is temperature (metal sinking-in processing temperature) within the limits exceeding 30-400 degrees C of melting points of the metal which sinks into the carbon material which performed the aforementioned heat treatment, and is the metal sinking-in method to the carbon material characterized by to sink in a metal below 1200 degrees C.

[0011] The contents of this invention are explained further in full detail below.

[0012] In this invention, it is calcinated at below the temperature that sinks in molten metal to the carbon material in the temperature requirement higher 30-400 degrees C than the melting point of the metal which sinks in as a carbon material used as a raw material, i.e., metal sinking-in processing temperature.

[0013] If it is going to sink in a metal, using such a carbon material as it is, since cracked gas will occur during sinking-in processing of a metal, it becomes difficult for a metal to advance into pore.

[0014] As a carbon material, to be a charge of the general electrode material manufactured from binding material, such as \*\*\*\* pitch coke, pitches, etc., such as a coal system and a petroleum system, or a raw coke, a special carbon material, and extruded material etc., not to limit especially, and what is necessary is just the carbon material with which a metal has the pore into which it may sink.

[0015] It is required to carry out a temperature up to heat treatment temperature under a non-oxidizing atmosphere on the occasion of heat treatment of the invention in this application. As a non-oxidizing atmosphere, they are inert gas atmosphere, such as vacuum atmosphere, argon gas, and nitrogen gas, for example.

[0016] In this invention, 900 degrees C or more less than 1200 degrees C of heat treatment temperature are preferably made comparatively into low temperature with 900-1100 degrees C, and heat treatment time is preferably set up more than for 30 minutes for 1 to 30 hours as for a long time [ it is more desirable and ] as 5 - 12 hours.

[0017] Since the heat treatment time for performing degasifying enough will become very long if heat treatment temperature cannot stop a degree of graphitization low above 1200 degrees C since graphitization speed becomes early as compared with the degasifying speed from a carbon material, and this temperature is less than 900 degrees C, it is not desirable.

[0018] Within the heat treatment temperature requirement of this invention, since graphitization will advance if it exceeds this, since degasifying will not fully be performed and degasifying will be mostly completed with heat treatment of 30 hours, if heat treatment time is less than 30 minutes, it is not desirable.

[0019] In order according to this method to be able to sink a metal into a low material of a degree of graphitization good, without raising the degree of graphitization of a carbon material since degasifying is possible, and to aim at improvement in a property, such as carburization-proof nature and abrasion resistance, and property control, such as lubricity, usefulness is high especially when it is necessary to stop the degree of graphitization of a carbon material low.

[0020] The degree of graphitization of a carbon material can compute and evaluate a size (Lc) or a spacing (d) of a microcrystal of C shaft orientations etc. from the result measured with the X-ray diffraction method.

[0021] Although not limited especially as a metal which sinks in, copper, a copper system alloy, lead, a lead system alloy, aluminum, an aluminum system alloy, tin, a tin system alloy, zinc, a zinc system alloy, etc. can be used. What is necessary is just to use these combining a carbon material according to the purpose of use.

[0022] The usual method is sufficient as the method of sinking in a metal into pore, for example, it inserts in a carbon material and a sinking-in metal in a high pressure vessel, as reduced pressure of about 0.1 or less Torrs, is immersed into the metal fused after fully deaerating in the gas in the pore of a carbon material, and should just switch the pressure in a high pressure vessel to pressurization using inert gas, such as nitrogen gas.

[0023] The metal for sinking in may be directly inserted in in a high pressure vessel, may dissolve, may prepare another container for the metal dissolution in the interior of a high pressure vessel, and may dissolve in the interior.

[0024] The temperature in this case is temperature higher 30 degrees C - 400 degrees C than the metaled melting point and desirable temperature high 150 degrees C - 300 degrees C, and is made into 1200 degrees C or less.

[0025] This has the bad fluidity of molten metal at under temperature higher 30 degrees C than the melting point, and since viscosity is high, it is because metaled sinking in is difficult.

[0026] Moreover, since metaled evaporation tends to take place by the process which carries out a temperature up and fuses a metal in the state of exceeding temperature higher 400 degrees C than the metaled melting point, metaled vapor pressure being high and carrying out deaeration processing of a carbon material when it reaches to the temperature greatly beyond the melting point, it is not desirable.

[0027] Moreover, it could be 1200 degrees C or less for suppressing graphitization as mentioned above.

[0028] The pressure at the time of pressurizing should just be a pressure higher than the value calculated using the surface tension of molten metal, and the pore diameter of a carbon material by the principle of capillarity.

[0029] Generally, when sinking for example, a bronze alloy (50:50 Cu:Sn= 90:10) into a carbon material with an average pore diameter of 4 micrometers, about two 50 kgf/cm are enough as the aforementioned pressure.

[0030] In order to make easy to take out the carbon material which carried out metal sinking in, a metal pulls up the carbon material with which it sank in from molten metal after fixed time progress in the state of pressurization.

[0031] Then, it can cool with the pressurization state in a high pressure vessel held, after a metal solidifies completely, it can decompress, and a metal can fully obtain the carbon material with which it sank in.

[0032]

[Example] Below, the example of this invention is shown. With rate of impregnation here, the rate of the volume which the metal with which it sank in among full open pore volume occupies is computed using bulk density change before and after sinking [ of a carbon material ] in, and metaled bulk density.

[0033] Moreover, porosity boiled the carbon material enough underwater and computed it by the Archimedes method.

[0034]

[Example 1] 1hr heat treatment was performed for the carbon material of the with a 1.46g [ /cm ] bulk density porosity [ 3 and 21% of porosity ] raw material calcinated at 900 degrees C at 1150 degrees C under argon atmosphere.

[0035] After loading with the dry bronze alloy (Cu:Sn=90:10) the carbon material after this heat treatment, and the carbon material whose melting point is 980 degrees C into the high pressure vessel and carrying out melting at 0.1Torr and 1050 degrees C, it was immersed, the carbon material was pressurized to 50 kg/cm<sup>2</sup> by argon gas, and it performed metal sinking in.

[0036] After holding in the state as it is for 1 hour successingly, when the carbon material was pulled up from molten metal, and

it cooled, holding the pressure in a high pressure vessel to 50 kg/cm<sup>2</sup> and the carbon-metal matrix composite was produced, metalate rate of impregnation was 80%.

[0037]

[Example 2] When 24hr heat treatment was performed at 1000 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 1, rate of impregnation was 76%.

[0038]

[Example 3] When heat treatment temperature was made into 1100 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 2, rate of impregnation was 79%.

[0039]

[Example 4] When heat treatment time was set to 12hr(s) and also the carbon-metal matrix composite was produced by the same method as an example 2, rate of impregnation was 77%.

[0040]

[Example 5] Using bulk density 1.43 g/cm<sup>3</sup> and the carbon material of 19% of porosity, 12hr(s) or 24hr heat treatment was performed at 1000 degrees C, and also the carbon-metal matrix composite was produced by the same method as an example 2.

[0041] Rate of impregnation was 83%, when 12hr heat treatment was carried out and 24hr heat treatment was carried out 76%.

[0042]

[The example 1 of comparison] When it did not heat-treat and also the carbon-metal matrix composite was produced by the same method as an example 1, rate of impregnation was 52%.

[0043]

[The example 2 of comparison] When it did not heat-treat and also the carbon-metal matrix composite was produced by the same method as an example 5, rate of impregnation was 50%.

[0044]

[The example 3 of comparison] Although rate of impregnation was 82% when it heat-treated at 1500 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 1, Lc of a carbon material was 5.814nm.

[0045] The result which measured and calculated the size Lc of the microcrystal of C shaft orientations of the carbon material prepared in the above-mentioned examples 1, 2, and 4 and the example 3 of comparison according to the X diffraction is the 1st table.

[0046] Although a degree of graphitization is hardly changing with the example 1 of comparison, as for the example 3 of comparison heat-treated at the elevated temperature, the examples 1, 2, and 4 which heat-treated at low temperature are known by that the degree of graphitization is high so that clearly from a table.

[0047]

[Table 1]

第 1 表

供試材	Lc (nm)
実施例 1	2. 5 2 1
実施例 2	2. 4 2 5
実施例 4	2. 2 9 6
比較例 1	2. 0 3 7
比較例 3	5. 8 1 4

[0048]

[Effect of the Invention] As mentioned above, according to this invention, even if it uses the carbon material calcinated below by the melting point of the metal which sinks in for a raw material, the carbon-metal matrix composite of very high rate of impregnation can be manufactured.

[0049] Moreover, the carbon-metal matrix composite of high rate of impregnation can be manufactured by heat-treating at low temperature comparatively for a long time, without hardly raising the degree of graphitization of a carbon material.

.....

[Translation done.]

\* NOTICES \*

**Japan Patent Office is not responsible for any damages caused by the use of this translation.**

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

---

CLAIMS

---

[Claim(s)]

[Claim 1] The metal sinking-in method to the carbon material characterized by to be temperature (metal sinking-in processing temperature) within the limits exceeding 30-400 degrees C of melting points of the metal which sinks in, and to sink a metal into the carbon material which performed the aforementioned heat treatment below 1200 degrees C after heat-treating more than for 30 minutes under a non-oxidizing atmosphere beforehand within the limits of 900-degree-C or more less than 1200-degree-C temperature to the carbon material calcinated below at the metal sinking-in processing temperature used as a raw material.

---

[Translation done.]

(19)



JAPANESE PATENT OFFICE

## PATENT ABSTRACTS OF JAPAN

(11) Publication number: 06263571 A

(43) Date of publication of application: 20.09.94

(51) Int. Cl

**C04B 41/88**  
**B60L 5/00**  
**C04B 35/52**

(21) Application number: 05077522

(22) Date of filing: 12.03.93

(71) Applicant: NIPPON STEEL CORP NIPPON STEEL CHEM CO LTD

(72) Inventor: FUJIMOTO KENICHIRO  
MUKAI KOICHIRO

**(54) METHOD FOR IMPREGNATING CARBON MATERIAL WITH METAL**

**(57) Abstract:**

**PURPOSE:** To improve the impregnation of a metal into a carbon material by impregnating the carbon material fired at a specific temperature with the metal melted at a higher temperature than the melting point of the metal in a nonoxidative atmosphere.

**CONSTITUTION:** A porous carbon material produced from petroleum pitch coke is subjected to a degassing treatment at 900-1200°C for 30min to 1-30hrs in a nonoxidative atmosphere such as Ar. A metal selected

from copper, copper alloy, aluminum, zinc, lead, etc., in response to the purpose is charged in a high pressure container, thermally melted at a temperature 30-400°C higher than the melting point of the metal but lower than 1200°C, and subsequently mixed with the treated carbon material under a high pressure to impregnate the carbon material with the melted metal. The carbon material is immersed in the melted metal for a prescribed time, discharged and subsequently cooled to provide the carbon-metal composite material having a high metal impregnation degree.

COPYRIGHT: (C)1994,JPO&amp;Japio

(19)日本国特許庁 (J-P)

(12)公開特許公報 (A)

(11)特許出願公開番号

特開平6-263571

(43)公開日 平成6年(1994)9月20日

(51) Int. Cl.<sup>5</sup>  
C 0 4 B 41/88  
B 6 0 L 5/00  
C 0 4 B 35/52

識別記号 V  
庁内整理番号 A 6821-5H  
G

F I

技術表示箇所

審査請求 未請求 請求項の数 1 FD (全 4 頁)

(21)出願番号 特願平5-77522

(22)出願日 平成5年(1993)3月12日

(71)出願人 000006655

新日本製鐵株式会社

東京都千代田区大手町2丁目6番3号

(71)出願人 000006644

新日鐵化学株式会社

東京都中央区銀座5丁目13番16号

(72)発明者 藤本 健一郎

川崎市中原区井田1618番地 新日本製鐵株式会社先端技術研究所内

(72)発明者 向井 幸一郎

川崎市中原区井田1618番地 新日本製鐵株式会社先端技術研究所内

(74)代理人 弁理士 井上 雅生

(54)【発明の名称】炭素材料への金属含浸方法

(57)【要約】

【目的】 金属含浸処理温度以下で焼成された炭素材料の気孔中に十分な量の金属を含浸する方法を提供する。また別の目的は、基材となる炭素材料の黒鉛化度をあまり上げることなく金属を含浸することである。

【構成】 原料となる金属含浸処理温度以下で焼成された炭素材料に、予め非酸化性雰囲気下で温度900℃以上1200℃未満の範囲内で30分間以上の熱処理を施すことにより、金属含浸処理中の炭素材料からのガス発生を抑制して金属含浸を良好に行なわせる。

## 【特許請求の範囲】

【請求項1】 原料となる金属含浸処理温度以下で焼成された炭素材料に、予め非酸化性雰囲気下で温度900℃以上1200℃未満の範囲内で30分間以上の熱処理を施した後、前記の熱処理を施した炭素材料に、含浸する金属の融点を30～400℃上回る温度（金属含浸処理温度）範囲内であって、1200℃以下で金属を含浸することを特徴とする炭素材料への金属含浸方法。

## 【発明の詳細な説明】

## 【0001】

【産業上の利用分野】本発明は、炭素材料の微細な気孔中に金属を溶融含浸して複合材料を製造する方法に関するものである。更に詳しくいえば、金属含浸処理温度以下で焼成された炭素材料へ金属含浸を良好に行わせる金属含浸方法に関するものである。

## 【0002】

【従来の技術】最近、炭素材料に金属を含浸して得られる複合材料は、摺動集電用材料、摺動材料等として注目を集めている。

【0003】通常、上記のような複合材料を製造する際の金属の含浸処理は、高圧容器内に金属と炭素材料を仕込んだ後、炭素材料の脱気処理を行いながら昇温して金属を溶融させ、溶融金属中に炭素材料を浸漬した状態で高圧容器内の圧力をあげ一定時間加圧含浸処理を施した後、炭素材料を溶融金属から引き上げて加圧状態を保持したまま降温する方法がとられている。

【0004】また、金属を溶融含浸する温度は、金属の含浸を容易にするために、金属の流動性を上げて粘度を下げることができるよう、含浸する金属の融点よりもかなり高温に設定されるのが一般的である。

【0005】しかし、金属をこれら炭素材料の微細な気孔中に十分に充填することは必ずしも容易ではなく、炭素材料の表面につながっている気孔であるにもかかわらず、金属が十分に含浸されない部分が少なからず発生しているのが現状である。

【0006】このため、炭素材料中の気孔への金属の十分な含浸を図る方法として、（1）炭素材料に予め金属塩の溶液を含浸し、熱処理して溶剤を除去することにより、溶融金属の炭素材料に対する濡れ性を改善する（特開昭61-136644号公報）、（2）炭素材料に対する濡れ性が良好な合金を用いる（特公昭52-822号公報）、（3）炭素材料を焼成炭化の温度以上の温度域で熱処理してから用いる（特開平4-83784号公報）等の方法が提案されている。

## 【0007】

【発明が解決しようとする課題】本発明の目的は、金属含浸処理温度以下で焼成された炭素材料中の気孔に十分な量の金属を含浸する方法を提供することにある。

【0008】また別の目的は、基材となる炭素材料の黒鉛化度をあまり上げることなく金属を含浸することである。

る。

## 【0009】

【課題を解決するための手段】本発明者らは、金属含浸処理に先立って炭素材料を熱処理して脱ガスして、金属含浸処理中の炭素材料からの分解ガスの発生を抑えるとともに、熱処理温度を抑制することにより、前記目的を達成することができることを見いだし、本発明を完成した。

【0010】本発明は、原料となる金属含浸処理温度以下で焼成された炭素材料に予め非酸化性雰囲気下で温度900℃以上1200℃未満の範囲内で30分間以上の熱処理を施した後、前記の熱処理を施した炭素材料に、含浸する金属の融点を30～400℃上回る温度（金属含浸処理温度）範囲内であって、1200℃以下で金属を含浸することを特徴とする炭素材料への金属含浸方法である。

【0011】以下に本発明の内容を更に詳述する。

【0012】本発明において、原料となる炭素材料としては、含浸する金属の融点より30～400℃高い温度範囲内の炭素材料へ溶融金属を含浸する温度、即ち金属含浸処理温度以下で焼成されたものである。

【0013】このような炭素材料を、そのまま用いて金属を含浸しようとすると、金属の含浸処理中に熱分解ガスが発生してしまうため、金属が気孔内に進入することが難しくなる。

【0014】炭素材料としては、石炭系、石油系等の煅焼ピッチコークスとピッチ等の結合材、或いは生コークス等から製造される、一般の電極材料、特殊炭素材料、押し出し材料等で、特に限定するものではなく、金属が含浸され得る気孔を有する炭素材料であればよい。

【0015】本願発明の熱処理に際しては、非酸化性雰囲気下で熱処理温度まで昇温することが必要である。非酸化性雰囲気としては、例えば真空雰囲気、アルゴンガス、窒素ガス等の不活性ガス雰囲気である。

【0016】本発明では、熱処理温度を900℃以上1200℃未満、好ましくは900～1100℃と比較的の低温とし、熱処理時間を30分間以上、好ましくは1～30時間、より好ましくは5～12時間と長めに設定する。

【0017】熱処理温度が1200℃以上では、炭素材料からの脱ガス速度と比較して黒鉛化速度が早くなるために黒鉛化度を低く抑えることができず、また該温度が900℃を下回ると脱ガスを充分行うための熱処理時間が極めて長くなるため好ましくない。

【0018】本発明の熱処理温度範囲内では、熱処理時間が30分を下回ると脱ガスが充分に行われず、また、脱ガスは30時間の熱処理でほぼ完了するのでこれを上回ると黒鉛化が進行してしまうため好ましくない。

【0019】この方法によれば、炭素材料の黒鉛化度を上げずに脱ガスが可能であるために、黒鉛化度の低い材

料に金属を良好に含浸することができ、耐摩耗性、耐摩耗性等の特性向上や、潤滑性などの特性制御を図るために、炭素材料の黒鉛化度を低く抑える必要がある際には特に有用性が高い。

【0020】炭素材料の黒鉛化度は、X線回折法により測定した結果からC軸方向の結晶子の大きさ(Lc)或いは面間隔(d)等を算出して評価できる。

【0021】含浸する金属としては特に限定されるものではないが、銅、銅系合金、鉛、鉛系合金、アルミニウム、アルミニウム系合金、錫、錫系合金、亜鉛、亜鉛系合金等が使用できる。これらは、使用目的に応じて、炭素材料と組み合わせて用いればよい。

【0022】金属を気孔内に含浸する方法は通常の方法でよく、例えば、高圧容器内に炭素材料、含浸金属を装入して、高圧容器内の圧力を0.1 Torr程度以下の減圧として炭素材料の気孔内のガスを十分に脱気した後、溶融した金属中に浸漬し、窒素ガス等の不活性ガスを用いて加圧に切り換えればよい。

【0023】含浸用の金属は、高圧容器内に直接装入して溶解してもよいし、高圧容器内部に金属溶解用の別の容器を設けてその内部で溶解してもよい。

【0024】この際の温度は、金属の融点より30℃～400℃高い温度、好ましくは150℃～300℃高い温度であって、1200℃以下とする。

【0025】これは、融点よりも30℃高い温度未満では溶融金属の流动性が悪く、粘度が高いため、金属の含浸が難しいためである。

【0026】また、金属の融点よりも400℃高い温度を越える状態では、金属の蒸気圧が高く、炭素材料の脱気処理をしながら昇温して金属を溶融する工程で、融点を大きく超えた温度まで到達した際に、金属の蒸発が起こり易いため好ましくない。

【0027】また、1200℃以下としたのは、前述のように黒鉛化を抑制するためである。

【0028】加圧する際の圧力は、毛細管現象の法則により溶融金属の表面張力と炭素材料の気孔径を用いて計算した値よりも高い圧力であればよい。

【0029】一般には、平均気孔径4μmの炭素材料に例えば青銅合金(Cu:Sn=50:50, 90:10)を含浸する場合、前記圧力は50kgf/cm<sup>2</sup>程度で十分である。

【0030】金属含浸した炭素材料を取り出し易くするために、加圧状態で一定時間経過後に、金属が含浸された炭素材料を溶融金属から引き上げる。

【0031】この後、高圧容器内の加圧状態を保持したまま冷却し、金属が完全に凝固してから減圧して、十分に金属が含浸された炭素材料を得ることができる。

【0032】

【実施例】以下に、本発明の実施例を示す。ここでいう含浸率とは、全開気孔体積のうちで含浸された金属が占

める体積の割合を、炭素材料の含浸前後での嵩密度変化と金属の嵩密度を用いて算出したものである。

【0033】また気孔率は、炭素材料を水中で十分煮沸して、アルキメデス法により算出した。

【0034】

【実施例1】900℃にて焼成した嵩密度1.46g/cm<sup>3</sup>、気孔率21%の原料の炭素材料を、アルゴン雰囲気下、1150℃で1hr熱処理を行った。

【0035】この熱処理後の炭素材料と融点が980℃の炭素材料には濡れない青銅合金(Cu:Sn=90:10)を高圧容器内に装填し、0.1Torr、1050℃で溶融させた後、炭素材料を浸漬し、アルゴンガスで50kgf/cm<sup>2</sup>まで加圧し金属含浸を行った。

【0036】引続き1時間そのままの状態に保持した後、炭素材料を溶融金属より引き上げて、高圧容器内の圧力を50kgf/cm<sup>2</sup>に保持したまま冷却し、炭素一金属複合材料を作製したところ、金属の含浸率は80%であった。

【0037】

【実施例2】1000℃で24hr熱処理を行うほかは実施例1と同様の方法で炭素一金属複合材料を作製したところ、含浸率は76%であった。

【0038】

【実施例3】熱処理温度を1100℃とするほかは実施例2と同様の方法で炭素一金属複合材料を作製したところ、含浸率は79%であった。

【0039】

【実施例4】熱処理時間を12hrとするほかは実施例2と同様の方法で炭素一金属複合材料を作製したところ、含浸率は77%であった。

【0040】

【実施例5】嵩密度1.43g/cm<sup>3</sup>、気孔率19%の炭素材料を用い、1000℃で12hr、或いは24hr熱処理を行うほかは実施例2と同様の方法で炭素一金属複合材料を作製した。

【0041】含浸率は12hr熱処理した場合76%、24hr熱処理した場合83%であった。

【0042】

【比較例1】熱処理を施さないほかは、実施例1と同様の方法で炭素一金属複合材料を作製したところ、含浸率は52%であった。

【0043】

【比較例2】熱処理を施さないほかは、実施例5と同様の方法で炭素一金属複合材料を作製したところ、含浸率は50%であった。

【0044】

【比較例3】1500℃にて熱処理を行うほかは実施例1と同様の方法で炭素一金属複合材料を作製したところ、含浸率は82%であったが、炭素材料のLcは5.814nmであった。

【0045】前述の実施例1、2、4、比較例3で準備した炭素材料のC軸方向の結晶子の大きさ  $L_c$  をX線回折により測定、計算した結果が第1表である。

【0046】表から明らかなように、低温で熱処理を行った実施例1、2、4は黒鉛化度がほとんど比較例1と変化していないが、高温で熱処理した比較例3は黒鉛化度が高くなっていることがわかる。

【0047】

【表1】

第1表

供試材	$L_c$ (nm)
実施例1	2.521
実施例2	2.425
実施例4	2.296
比較例1	2.037
比較例3	5.814

【0048】

【発明の効果】以上のように、本発明によれば、含浸する金属の融点以下で焼成された炭素材料を原料に用いても、極めて高含浸率の炭素-金属複合材料を製造することができる。

【0049】また、比較的低温で長時間熱処理することにより、炭素材料の黒鉛化度をほとんど上げずに、高含浸率の炭素-金属複合材料を製造することができる。

# PATENT ABSTRACTS OF JAPAN

(11)Publication number : 06-263571

(43)Date of publication of application : 20.09.1994

---

(51)Int.Cl.

C04B 41/88  
B60L 5/00  
C04B 35/52

---

(21)Application number : 05-077522

(71)Applicant : NIPPON STEEL CORP  
NIPPON STEEL CHEM CO LTD

(22)Date of filing : 12.03.1993

(72)Inventor : FUJIMOTO KENICHIRO  
MUKAI KOICHIRO

---

## (54) METHOD FOR IMPREGNATING CARBON MATERIAL WITH METAL

### (57)Abstract:

**PURPOSE:** To improve the impregnation of a metal into a carbon material by impregnating the carbon material fired at a specific temperature with the metal melted at a higher temperature than the melting point of the metal in a nonoxidative atmosphere.

**CONSTITUTION:** A porous carbon material produced from petroleum pitch coke is subjected to a degassing treatment at 900-1200°C for 30min to 1-30hrs in a nonoxidative atmosphere such as Ar. A metal selected from copper, copper alloy, aluminum, zinc, lead, etc., in response to the purpose is charged in a high pressure container, thermally melted at a temperature 30-400°C higher than the melting point of the metal but lower than 1200°C, and subsequently mixed with the treated carbon material under a high pressure to impregnate the carbon material with the melted metal. The carbon material is immersed in the melted metal for a prescribed time, discharged and subsequently cooled to provide the carbon-metal composite material having a high metal impregnation degree.

---

## LEGAL STATUS

[Date of request for examination]

[Date of sending the examiner's decision of rejection]

[Kind of final disposal of application other than the examiner's decision of rejection or

## \* NOTICES \*

**Japan Patent Office is not responsible for any damages caused by the use of this translation.**

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

## DETAILED DESCRIPTION

## [Detailed Description of the Invention]

[0001]

[Industrial Application] this invention relates to the method of carrying out melting sinking in of the metal into the detailed pore of a carbon material, and manufacturing composite material. Furthermore, if it says in detail, it is related with the metal sinking-in method of making it performing good to the carbon material calcinated below at metal sinking-in processing temperature to metal sink in.

[0002]

[Description of the Prior Art] Recently, the composite material sunk in and obtained by the carbon material in a metal attracts attention as the charge of sliding current collection material, a sliding material, etc.

[0003] Usually, the method of lowering pulling up a carbon material from molten metal, and holding a pressurization state, after having carried out the temperature up, having carried out melting of the metal, raising the pressure in a high pressure vessel with the state where the carbon material was immersed into molten metal and performing fixed time pressurization sinking-in processing, while sinking-in processing of the metal at the time of manufacturing the above composite material performed deaeration processing of a carbon material, after preparing the metal and the carbon material in the high pressure vessel is taken.

[0004] Moreover, in order to make it easy metaled to sink in, being set as a remarkable elevated temperature is more common [ the temperature which carries out melting sinking in of the metal ] than the melting point of the metal which sinks in so that a metaled fluidity may be raised and viscosity can be lowered.

[0005] However, in spite of it not being necessarily easy to fully be filled up with a metal into the detailed pore of these carbon materials and being the pore connected on the surface of the carbon material, the present condition is that the portion into which a metal does not fully sink has occurred not a little.

[0006] For this reason, the thing for which the solution of a metal salt is beforehand sunk in and heat-treated to (1) carbon material as a method of aiming at sufficient sinking [ of the metal to the pore in a carbon material ] in, and a solvent is removed, Methods, such as using, after the wettability to (2) carbon materials which improves the wettability to the carbon material of molten metal (Provisional-Publication-No. No. 136644 [ 61 to ] official report) heat-treats (3) carbon materials using a good alloy (Japanese Patent Publication No. No. 822 [ 52 to ] official report) in the temperature region more than the temperature of baking carbonization (publication-number No. 83784 [ four to ] official report), are proposed.

[0007]

[Problem(s) to be Solved by the Invention] The purpose of this invention is to offer the method of sinking in the metal of sufficient amount for the pore in the carbon material calcinated below at metal sinking-in processing temperature.

[0008] Moreover, another purpose is sinking in a metal, without raising the degree of graphitization of

the carbon material used as a base material not much.

[0009]

[Means for Solving the Problem] By suppressing heat treatment temperature, this invention persons found out that the aforementioned purpose could be attained, and completed this invention while they heat-treated and did degasifying of the carbon material in advance of metal sinking-in processing and suppressed generating of the cracked gas from the carbon material under metal sinking-in processing.

[0010] After this invention heat-treats more than for 30 minutes under a non-oxidizing atmosphere beforehand within the limits of 900 degrees-C or more less than 1200 degree-C temperature to the carbon material calcinated below at the metal sinking-in processing temperature used as a raw material, it is temperature (metal sinking-in processing temperature) within the limits exceeding 30-400 degrees C of melting points of the metal which sinks into the carbon material which performed the aforementioned heat treatment, and is the metal sinking-in method to the carbon material characterized by to sink in a metal below 1200 degrees C.

[0011] The contents of this invention are explained further in full detail below.

[0012] In this invention, it is calcinated at below the temperature that sinks in molten metal to the carbon material in the temperature requirement higher 30-400 degrees C than the melting point of the metal which sinks in as a carbon material used as a raw material, i.e., metal sinking-in processing temperature.

[0013] If it is going to sink in a metal, using such a carbon material as it is, since cracked gas will occur during sinking-in processing of a metal, it becomes difficult for a metal to advance into pore.

[0014] As a carbon material, to be a charge of the general electrode material manufactured from binding material, such as \*\*\*\* pitch coke, pitches, etc., such as a coal system and a petroleum system, or a raw coke, a special carbon material, and extruded material etc., not to limit especially, and what is necessary is just the carbon material with which a metal has the pore into which it may sink.

[0015] It is required to carry out a temperature up to heat treatment temperature under a non-oxidizing atmosphere on the occasion of heat treatment of the invention in this application. As a non-oxidizing atmosphere, they are inert gas atmosphere, such as vacuum atmosphere, argon gas, and nitrogen gas, for example.

[0016] In this invention, 900 degrees C or more less than 1200 degrees C of heat treatment temperature are preferably made comparatively into low temperature with 900-1100 degrees C, and heat treatment time is preferably set up more than for 30 minutes for 1 to 30 hours as for a long time [ it is more desirable and ] as 5 - 12 hours.

[0017] Since the heat treatment time for performing degasifying enough will become very long if heat treatment temperature cannot stop a degree of graphitization low above 1200 degrees C since graphitization speed becomes early as compared with the degasifying speed from a carbon material, and this temperature is less than 900 degrees C, it is not desirable.

[0018] Within the heat treatment temperature requirement of this invention, since graphitization will advance if it exceeds this, since degasifying will not fully be performed and degasifying will be mostly completed with heat treatment of 30 hours, if heat treatment time is less than 30 minutes, it is not desirable.

[0019] In order according to this method to be able to sink a metal into a low material of a degree of graphitization good, without raising the degree of graphitization of a carbon material since degasifying is possible, and to aim at improvement in a property, such as carburization-proof nature and abrasion resistance, and property control, such as lubricity, usefulness is high especially when it is necessary to stop the degree of graphitization of a carbon material low.

[0020] The degree of graphitization of a carbon material can compute and evaluate a size (Lc) or a spacing (d) of a microcrystal of C shaft orientations etc. from the result measured with the X-ray diffraction method.

[0021] Although not limited especially as a metal which sinks in, copper, a copper system alloy, lead, a lead system alloy, aluminum, an aluminum system alloy, tin, a tin system alloy, zinc, a zinc system alloy, etc. can be used. What is necessary is just to use these combining a carbon material according to the purpose of use.

[0022] The usual method is sufficient as the method of sinking in a metal into pore, for example, it inserts in a carbon material and a sinking-in metal in a high pressure vessel, as reduced pressure of about 0.1 or less Torrs, is immersed into the metal fused after fully deaerating in the gas in the pore of a carbon material, and should just switch the pressure in a high pressure vessel to pressurization using inert gas, such as nitrogen gas.

[0023] The metal for sinking in may be directly inserted in in a high pressure vessel, may dissolve, may prepare another container for the metal dissolution in the interior of a high pressure vessel, and may dissolve in the interior.

[0024] The temperature in this case is temperature higher 30 degrees C - 400 degrees C than the metaled melting point and desirable temperature high 150 degrees C - 300 degrees C, and is made into 1200 degrees C or less.

[0025] This has the bad fluidity of molten metal at under temperature higher 30 degrees C than the melting point, and since viscosity is high, it is because metaled sinking in is difficult.

[0026] Moreover, since metaled evaporation tends to take place by the process which carries out a temperature up and fuses a metal in the state of exceeding temperature higher 400 degrees C than the metaled melting point, metaled vapor pressure being high and carrying out deaeration processing of a carbon material when it reaches to the temperature greatly beyond the melting point, it is not desirable.

[0027] Moreover, it could be 1200 degrees C or less for suppressing graphitization as mentioned above.

[0028] The pressure at the time of pressurizing should just be a pressure higher than the value calculated using the surface tension of molten metal, and the pore diameter of a carbon material by the principle of capillarity.

[0029] Generally, when sinking for example, a bronze alloy (50:50 Cu:Sn= 90:10) into a carbon material with an average pore diameter of 4 micrometers, about two 50 kgf/cm are enough as the aforementioned pressure.

[0030] In order to make easy to take out the carbon material which carried out metal sinking in, a metal pulls up the carbon material with which it sank in from molten metal after fixed time progress in the state of pressurization.

[0031] Then, it can cool with the pressurization state in a high pressure vessel held, after a metal solidifies completely, it can decompress, and a metal can fully obtain the carbon material with which it sank in.

[0032]

[Example] Below, the example of this invention is shown. With rate of impregnation here, the rate of the volume which the metal with which it sank in among full open pore volume occupies is computed using bulk density change before and after sinking [ of a carbon material ] in, and metaled bulk density.

[0033] Moreover, porosity boiled the carbon material enough underwater and computed it by the Archimedes method.

[0034]

[Example 1] 1hr heat treatment was performed for the carbon material of the with a 1.46g [/cm ] bulk density porosity [ 3 and 21% of porosity ] raw material calcinated at 900 degrees C at 1150 degrees C under argon atmosphere.

[0035] After loading with the dry bronze alloy (Cu:Sn=90:10) the carbon material after this heat treatment, and the carbon material whose melting point is 980 degrees C into the high pressure vessel and carrying out melting at 0.1Torr and 1050 degrees C, it was immersed, the carbon material was pressurized to 50 kg/cm<sup>2</sup> by argon gas, and it performed metal sinking in.

[0036] After holding in the state as it is for 1 hour successingly, when the carbon material was pulled up from molten metal, and it cooled, holding the pressure in a high pressure vessel to 50 kg/cm<sup>2</sup> and the carbon-metal matrix composite was produced, metaled rate of impregnation was 80%.

[0037]

[Example 2] When 24hr heat treatment was performed at 1000 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 1, rate of impregnation was 76%.

[0038]

[Example 3] When heat treatment temperature was made into 1100 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 2, rate of impregnation was 79%.  
[0039]

[Example 4] When heat treatment time was set to 12hr(s) and also the carbon-metal matrix composite was produced by the same method as an example 2, rate of impregnation was 77%.  
[0040]

[Example 5] Using bulk density 1.43 g/cm<sup>3</sup> and the carbon material of 19% of porosity, 12hr(s) or 24hr heat treatment was performed at 1000 degrees C, and also the carbon-metal matrix composite was produced by the same method as an example 2.

[0041] Rate of impregnation was 83%, when 12hr heat treatment was carried out and 24hr heat treatment was carried out 76%.

[0042]

[The example 1 of comparison] When it did not heat-treat and also the carbon-metal matrix composite was produced by the same method as an example 1, rate of impregnation was 52%.

[0043]

[The example 2 of comparison] When it did not heat-treat and also the carbon-metal matrix composite was produced by the same method as an example 5, rate of impregnation was 50%.

[0044]

[The example 3 of comparison] Although rate of impregnation was 82% when it heat-treated at 1500 degrees C and also the carbon-metal matrix composite was produced by the same method as an example 1, Lc of a carbon material was 5.814nm.

[0045] The result which measured and calculated the size Lc of the microcrystal of C shaft orientations of the carbon material prepared in the above-mentioned examples 1, 2, and 4 and the example 3 of comparison according to the X diffraction is the 1st table.

[0046] Although a degree of graphitization is hardly changing with the example 1 of comparison, as for the example 3 of comparison heat-treated at the elevated temperature, the examples 1, 2, and 4 which heat-treated at low temperature are known by that the degree of graphitization is high so that clearly from a table.

[0047]

[Table 1]

第 1 表

供試材	L c (nm)
実施例 1	2. 5 2 1
実施例 2	2. 4 2 5
実施例 4	2. 2 9 6
比較例 1	2. 0 3 7
比較例 3	5. 8 1 4

[0048]

[Effect of the Invention] As mentioned above, according to this invention, even if it uses the carbon material calcinated below by the melting point of the metal which sinks in for a raw material, the carbon-metal matrix composite of very high rate of impregnation can be manufactured.

[0049] Moreover, the carbon-metal matrix composite of high rate of impregnation can be manufactured by heat-treating at low temperature comparatively for a long time, without hardly raising the degree of graphitization of a carbon material.

**\* NOTICES \***

**Japan Patent Office is not responsible for any damages caused by the use of this translation.**

1. This document has been translated by computer. So the translation may not reflect the original precisely.
2. \*\*\* shows the word which can not be translated.
3. In the drawings, any words are not translated.

---

**CLAIMS**

---

[Claim(s)]

[Claim 1] The metal sinking-in method to the carbon material characterized by to be temperature (metal sinking-in processing temperature) within the limits exceeding 30-400 degrees C of melting points of the metal which sinks in, and to sink a metal into the carbon material which performed the aforementioned heat treatment below 1200 degrees C after heat-treating more than for 30 minutes under a non-oxidizing atmosphere beforehand within the limits of 900-degree-C or more less than 1200-degree-C temperature to the carbon material calcinated below at the metal sinking-in processing temperature used as a raw material.

---

[Translation done.]